ISOTOPIC MEASUREMENTS OF MURCHISON AND SEMARKONA RESIDUES BY TOF-SIMS. A. J. Fahey[1] and S. Messenger[2], [1]NIST, 100 Bureau Dr. Stop 8371 Gaithersburg, MD 20899-8371 (albert.fahey@nist.gov), [2] NIST, 100 Bureau Dr. Stop 8371 Gaithersburg, MD 20899-8371 (scott.messenger@nist.gov)

Introduction: We have developed a method to measure isotopic ratios in individual particles by Time of Flight Secondary Ion Mass Spectrometry (ToF-SIMS). ToF-SIMS is capable of achieving high spatial resolution (~200 nm beam diameter) with a simultaneous mass resolution high enough to "resolve" hydrides at the masses of elements such as Mg, Si, and Ti. In addition, ToF-SIMS consumes very little material during analysis and collects data at all masses from the particle under analysis. Consequently, the data rate is lower than with magnetic sector SIMS, but for some problems this is an acceptable trade-off. We have measured numerous terrestrial materials for their Si and Mg isotopic ratios as well as individual grains from Murchison and Semarkona.

Technique: The data reported here were take with a Cameca/IonTof ToF-SIMS IV[1]. Typically a Ga field-emission ion source is used to generate secondary ions for analysis. This instrument has the capability of running the Ga source in a configuration known as "burst" mode. In "burst" mode bursts of 1.2 ns primary ion pulses hit the sample at ~25 ns intervals producing a string of pulses for each mass. The advantages of this mode are that it produces a high spatial resolution beam (~200 nm) and a high mass resolution spectrum. In addition the "pulse string" increases the secondary ion data rate.

In order to make isotopic ratio measurements an algorithm was developed and a program written to properly combine the "burst" peaks into a single peak, perform the correct integration and interference stripping where appropriate. Hydride peak stripping was performed for the ^{29}Si and ^{25}Mg data reported here. A dead time correction of 70 ns was made to the data according to previously described methods[2].

Data: Spectra were obtained for ~50 terrestrial SiC particles, ~40 Murchison SiC particles and ~10 from Semarkona. Figure 1 shows the Si isotope data for all of the particles measured. The data are not corrected for mass fractionation in order to show that the instrumental mass fractionation is reasonable. Figure 2 shows a detail of the region around the origin of Figure 1.

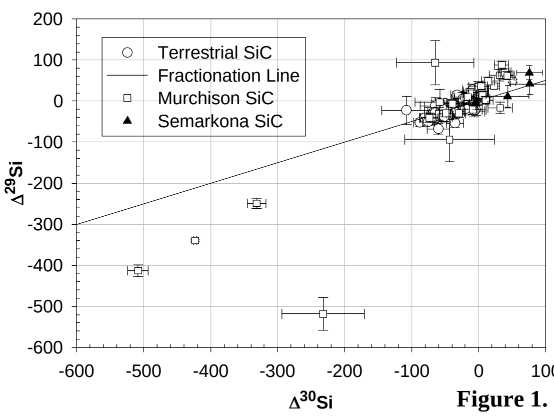

Figure 1.

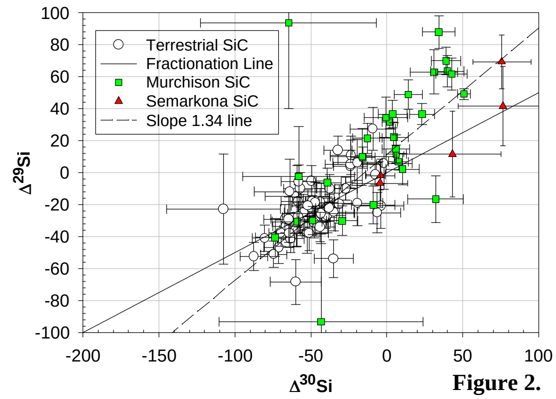

Figure 2.

Figure 2 shows that the data for the terrestrial SiC cluster around the mass fractionation line and have a weighted mean Δ^{29}Si= -22.3±1.3 ‰ (χ^2_ν = 3.2) and Δ^{30}Si= -47.2±1.4 ‰ (χ^2_ν = 3.8).

The Mg isotopic data for the standards (Burma spinel) and the Murchison SiC are plotted in Figure 3. All of the Burma spinel data plotted along the mass fractionation line and are hidden in a spot at the origin of Figure 3. The ^{26}Mg abundances from Murchison shows the greatest range of variation as expected. The Mg isotope signals in the Semarkona particles were either overwhelmed by background or were of terrestrial composition.

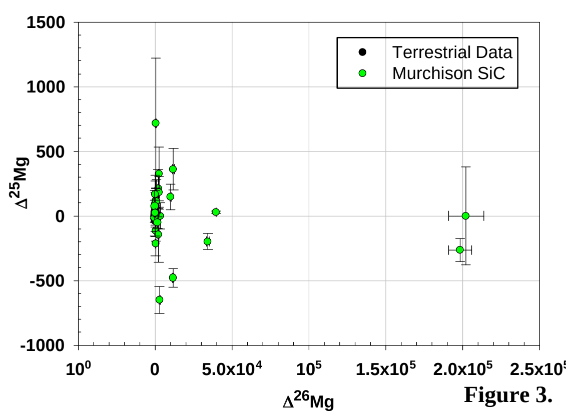

Figure 3.

The Si isotopic compositions of the Semarkona particles lie along the "mainstream" SiC line of slope 1.34[3] as do most of the Murchison grains. Of the ~40 Murchison SiC grains measured 4 X-grains were discovered. After the first X-grain was discovered the Mg isotopic ratios were extracted from the spectrum showing a large excess in ^{26}Mg from the decay of ^{26}Al. The ^{26}Mg signal was strong enough to allow us to image at 26 u to identify X-grains immediately when performing routine searching for analysis.

One X-grain that was discovered allowed us to exploit the high spatial resolution of the ToF-SIMS since it was directly adjacent to a "mainstream" SiC. Figure 4 shows the ion images of ^{26}Mg, ^{27}Al, ^{28}Si and all other ions from the Murchison SiC mount in the region of one of the X-grains. The ^{26}Mg ion image shows a small region of enriched ^{26}Mg. When compared to the ^{28}Si image one can see a single extended "bright" region in the same general position of the enriched ^{26}Mg. In fact, the extended region is two SiC grains, each ~1µm in size, touching each other. We were able to measure each separately. The Si and Mg isotopic composition of the X-grain (the lower portion of the extended region) is: Δ^{29}Si = -249±13 ‰, Δ^{30}Si = -331±14 ‰, Δ^{25}Mg = -264±91 ‰, Δ^{26}Mg = 198268±7551 ‰ while the adjacent "mainstream" SiC grain has: Δ^{29}Si = 15±9 ‰, Δ^{30}Si = 6±9 ‰, Δ^{25}Mg = -53±44 ‰, Δ^{26}Mg = 1524±76 ‰. We have not yet determined the degree of cross-contamination in the signals however the Mg isotopic composition sets a lower limit of a factor of ~100 on the "isotopic crosstalk" at this particle separation.

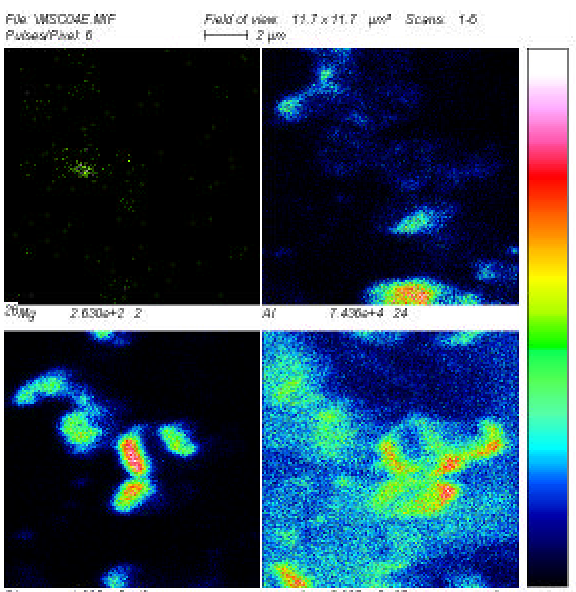

Figure 4.

Additional data exist in the spectra collected but it has yet to be extracted and quantified. Many of the SiC particles have significant signals from elements such as Ca, Ti, Cr, and Fe. However, standards must be measured and methodologies prepared to resolve interferences and produce the correct isotopic ratios.

Conclusions: Isotopic measurements of Si and Mg have been successfully performed for small (~1 µm) particles in both standards and meteoritic material. Four X-grains were discovered out of ~40 measured SiC grains from Murchison. This is a relatively high fraction according to the current body of data that exists on the meteoritic SiC[3]. One of the X-grains was in intimate contact with a "mainstream" SiC grain leading one to speculate that we may be able to find more of these unusual grains in the ToF-SIMS that may have been hidden to the magnetic sector SIMS instruments.

The ToF-SIMS generates a large amount of data for each particle measured. As yet we have not finished developing programs that will allow us to sort through the data in a reasonable amount of time. In addition we have plans to automate location and measurement of particles so that data from many more particles can be obtained. Automation may shed new light on the characteristics of the particle populations since the entire mass spectrum is collected for each grain.

References: [1] Certain commercial equipment, instruments, or materials are identified in this paper in order to adequately specify the experimental procedure. Such identification does not imply recommendation or endorsement by the National Institute of Standards and Technology, nor does it imply that the materials or equipment identified are necessarily the best available for the purpose. [2] Stephan T. et al (1994) *J. Vac. Sci Technol., 12(2)*, 405-410. [3] Hoppe P. and Ott U (1997) Astrophysical Implications of the Laboratory Study of Presolar Materials, AIP, Woodbury, New York, 27-58.